Detection of *p*-process ^{146}Sm nuclide by accelerator mass spectrometry

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Tb146 8 s 1+ * EC	Tb147 1.7 h (1/2+) * EC	Tb148 60 m 2- * EC	Tb149 4.118 h 1/2+ * EC,α	Tb150 3.48 h (2-) * EC,α	Tb151 17.609 h 1/2(+) * EC,α	Tb152 17.5 h 2- * EC,α	Tb153 2.34 d 5/2+ EC	Tb154 21.5 h 0 * EC,β-	Tb155 5.32 d 3/2+ EC	Tb156 5.35 d 3- * EC,β-	Tb157 71 y 3/2+ EC	Tb158 180 y 3- * EC,β-
Gd145 23.0 m 1/2+ * EC	Gd146 48.27 d 0+ EC	Gd147 38.06 h 7/2- EC	Gd148 74.6 y 0+ α	Gd149 9.28 d 7/2- EC,α	Gd150 1.79E6 y 0+ α	Gd151 124 d 7/2- EC,α	Gd152 1.08E14 y 0+ α 0.20	Gd153 241.6 d 3/2- EC	Gd154 0+ 2.18	Gd155 3/2- * 14.80	Gd156 0+ 20.47	Gd157 3/2- 15.65
Eu144 10.2 s 1+ EC	Eu145 5.93 d 5/2+ EC	Eu146 4.59 d 4- EC	Eu147 24.1 d 5/2+ EC,α	Eu148 54.5 d 5- EC,α	Eu149 93.1 d 5/2+ EC	Eu150 36.9 y 5(-) * EC	Eu151 5/2+ 47.8	Eu152 13.537 y 3- * EC,β-	Eu153 5/2+ 52.2	Eu154 8.593 y 3- * EC,β-	Eu155 4.7611 y 5/2+ β-	Eu156 15.19 d 0+ β-
Sm143 8.83 m 3/2+ * EC	Sm144 0+ 3.1	Sm145 340 d 7/2- EC	Sm146 1.03E+8 y 0+ α	Sm147 1.06E+11 y 7/2- α 15.0	Sm148 7E+15 y 0+ α 11.3	Sm149 2E+15 y 7/2- 13.8	Sm150 0+ 7.4	Sm151 90 y 5/2- β-	Sm152 0+ 26.7	Sm153 46.27 h 3/2+ * β-	Sm154 0+ 22.7	Sm155 22.3 m 3/2- β-
Pm142 40.5 s 1+ * EC	Pm143 265 d 5/2+ EC	Pm144 363 d 5- EC	Pm145 17.7 y 5/2+ EC,α	Pm146 5.53 y 3- EC,β-	Pm147 2.6234 y 7/2+ β-	Pm148 5.370 d 1- * β-	Pm149 53.08 h 7/2+ β-	Pm150 2.68 h (1-) β-	Pm151 28.40 h 5/2+ β-	Pm152 4.12 m 1+ * β-	Pm153 5.4 m 5/2- β-	Pm154 1.73 m (0,1) * β-
Nd141 2.49 h 3/2+ * EC	Nd142 0+ 27.13	Nd143 7/2- 12.18	Nd144 2.29E+15 y 0+ α 23.80	Nd145 7/2- 8.30	Nd146 0+ 17.19	Nd147 10.98 d 5/2- β-	Nd148 0+ 5.76	Nd149 1.728 h 5/2- β-	Nd150 1.1E19 y 0+ β- 5.64	Nd151 12.44 m (3/2)+ β-	Nd152 11.4 m 0+ β-	Nd153 28.9 s (3/2-) β-

r

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Gd145 23.0 m 1/2+ EC	Gd146 48.27 d 0+ EC	Gd147 38.06 h 7/2- EC	Gd148 74.6 y 0+ α	Gd149 9.28 d 7/2- EC,α	Gd150 1.79E6 y 0+ α	Gd151 124 d 7/2- EC,α	Gd152 1.08E14 y 0+ α 0.20	Gd153 241.6 d 3/2- EC	Gd154 0+ 2.18	Gd155 3/2- 14.80	Gd156 0+ 20.47	Gd157 3/2- 15.65
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S.B. Jacobsen, G.J. Wasserburg, Earth Planet. Sci. Lett. 67, 137 (1984):
$^{143}Nd - ^{147}Sm$ isochron in St-Severin meteorite : t = 4.55 ± 0.33 Gy

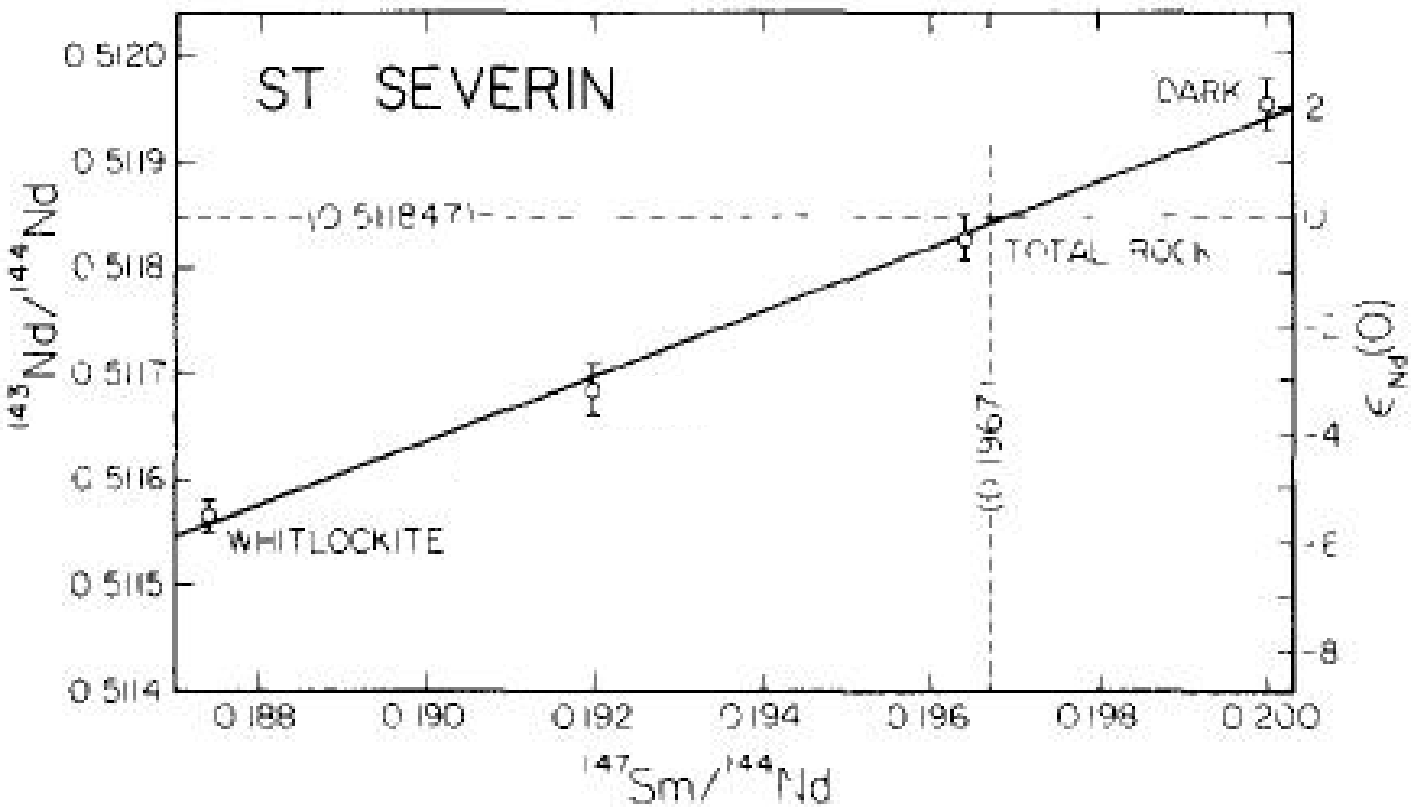


Fig 3 Sm-Nd evolution diagram for the St Severin chondrite

S.B. Jacobsen, G.J. Wasserburg, Earth Planet. Sci. Lett. 67, 137 (1984):
^{146}Sm in-situ decay in meteorites : ^{146}Sm present in Early-Solar system

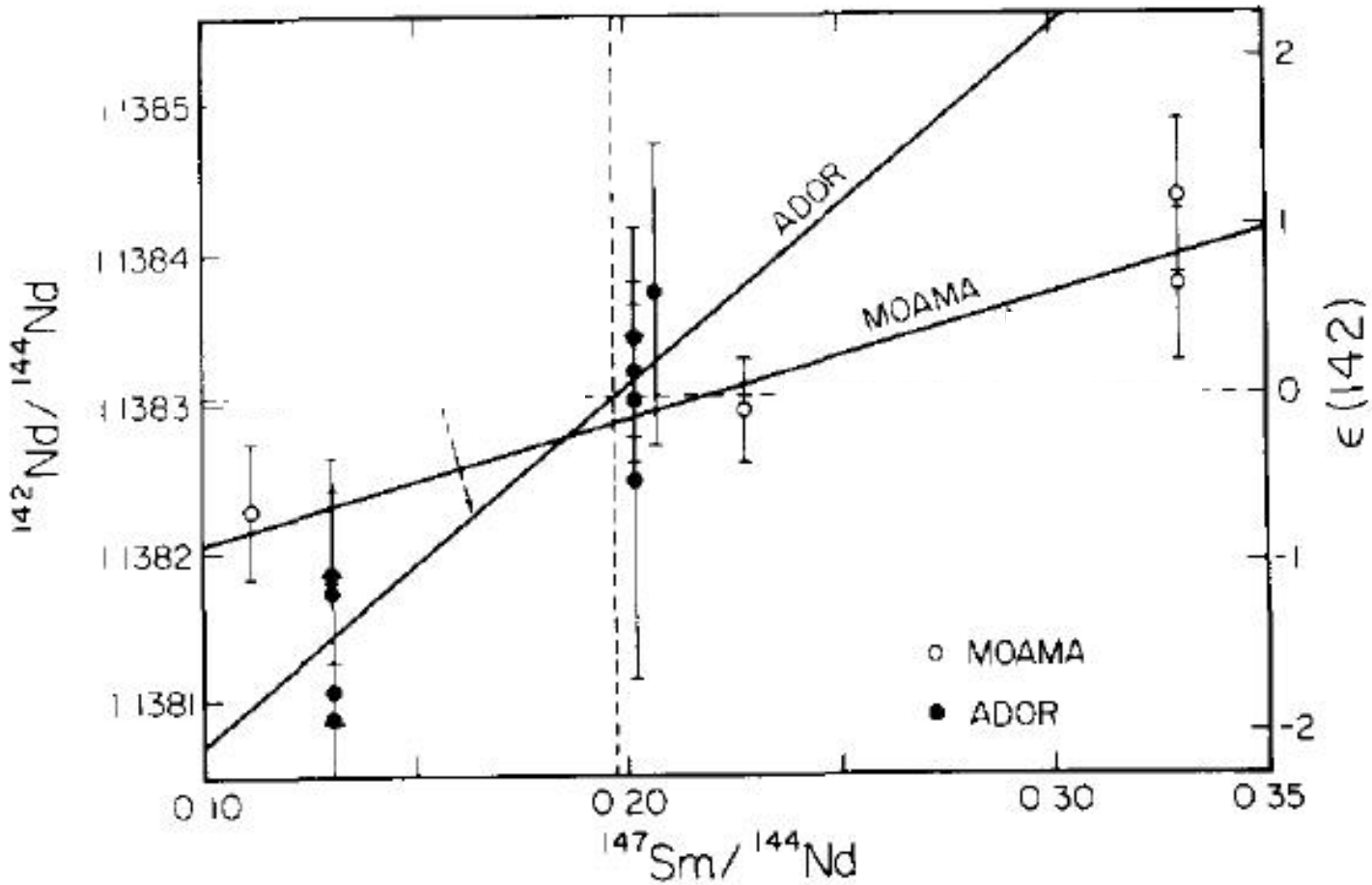
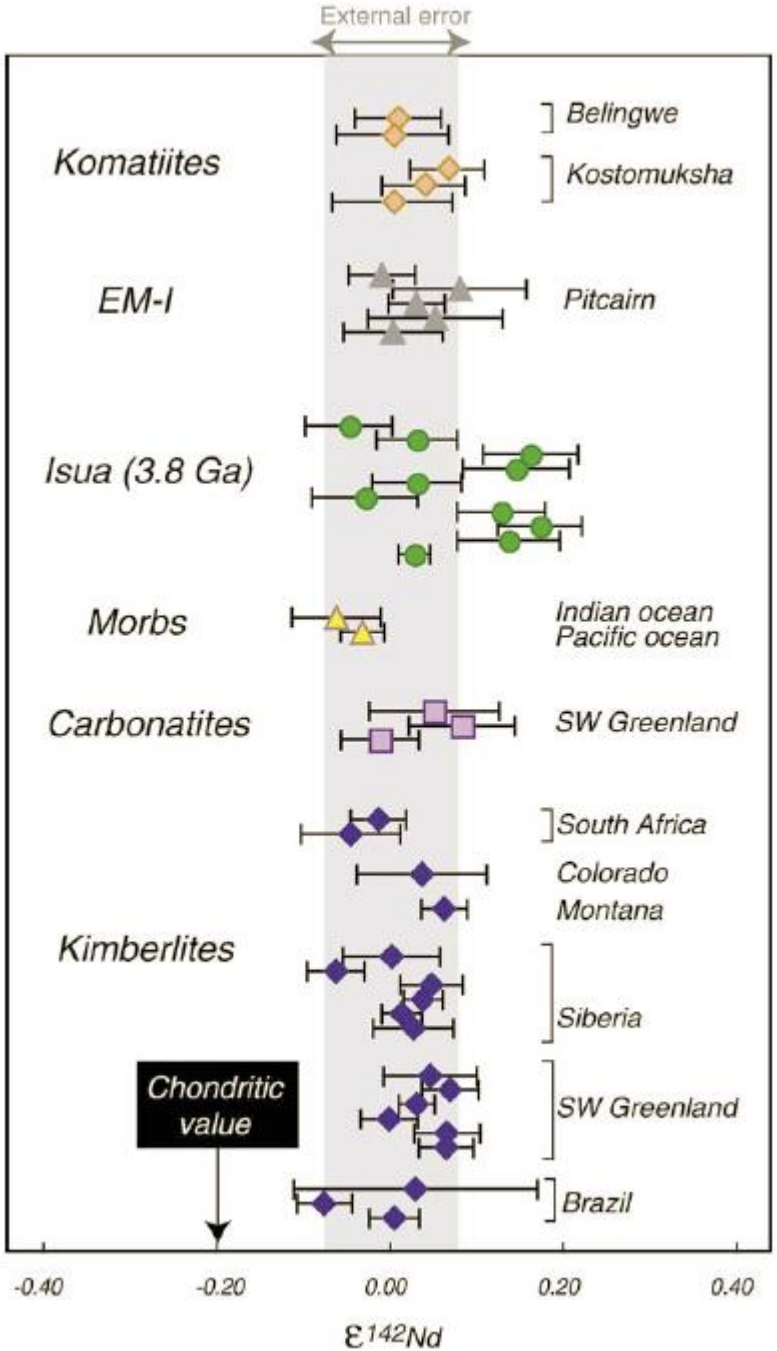


Fig 6 ^{142}Nd/^{144}Nd evolution diagram for the achondrites Moama and Angra dos Reis.



M. Boyet and R.W. Carlson, EPSL 250, 254 (2006)

$\varepsilon(^{142}Nd)$: deviation in parts per 10,000

Possible scenarios:

1. Earth condensed from non-chondritic material
2. Early differentiation of Sm-rich material during planetary formation: Earth should therefore contain a complementary reservoir of low Sm/Nd ratio.

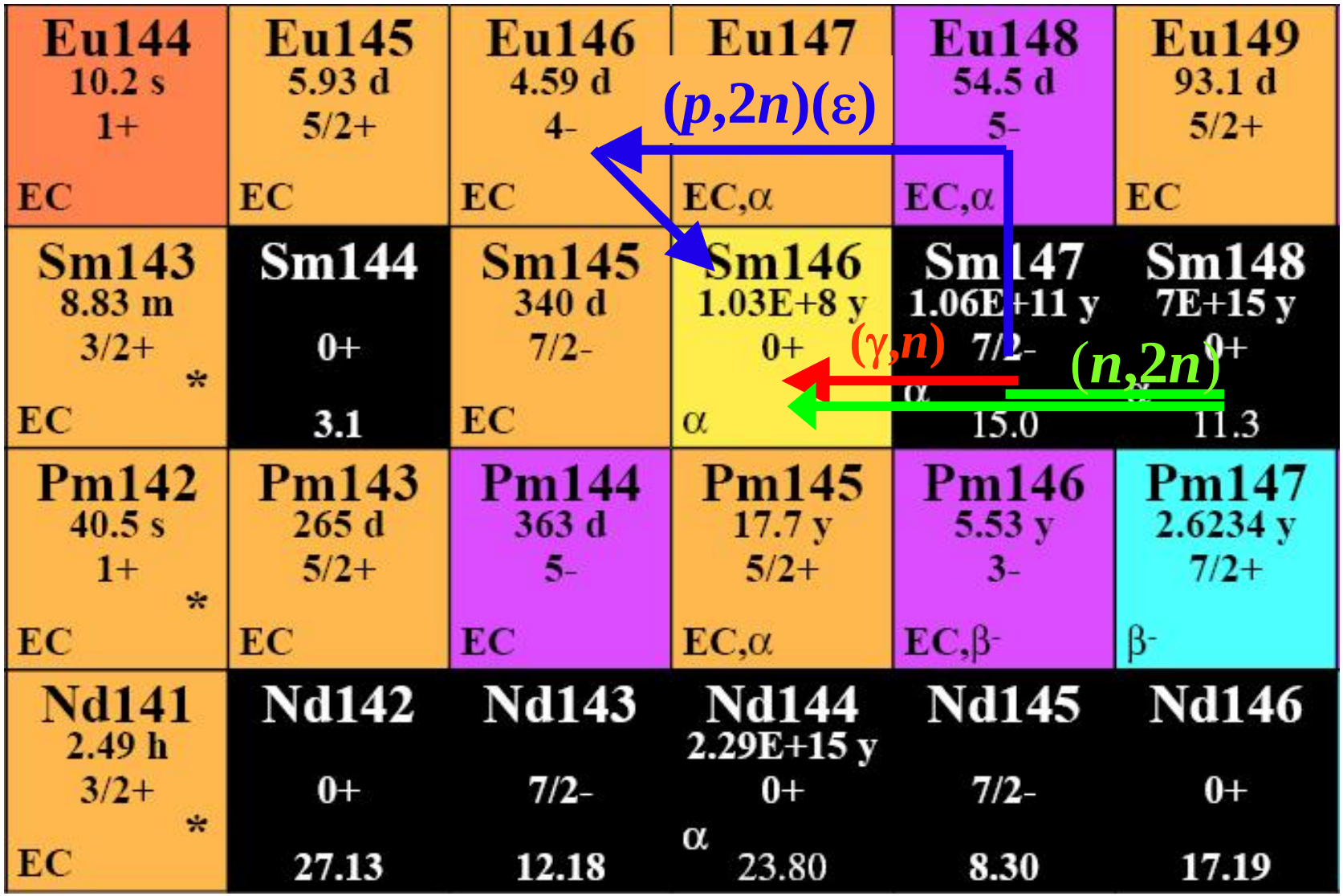
Direct detection of ^{146}Sm by accelerator mass spectrometry: the interest

1. Possible determination of cross section of ^{142}Nd(α,γ)^{146}Sm at low energy by direct counting for study of the inverse (γ,α) p-process reaction

2. Re-determination of ^{146}Sm half-life, important for Early-Solar System studies and planet formation

3. ^{146}Sm in nature:
   - cosmogenic isotope : ^{147}Sm(n,2n) in meteorites ?
   - accretion from interstellar medium ?

Laboratory production of ^{146}Sm



AMS detection technique:

1. Use of ECR ion source and ATLAS to reach high energy

2. Use of Gas-Filled Magnet to separate ^{146}Sm and ^{146}Nd

Eu144 10.2 s 1+ EC	Eu145 5.93 d 5/2+ EC	Eu146 4.59 d 4- EC	Eu147 24.1 d 5/2+ EC,α	Eu148 54.5 d 5- EC,α	Eu149 93.1 d 5/2+ EC
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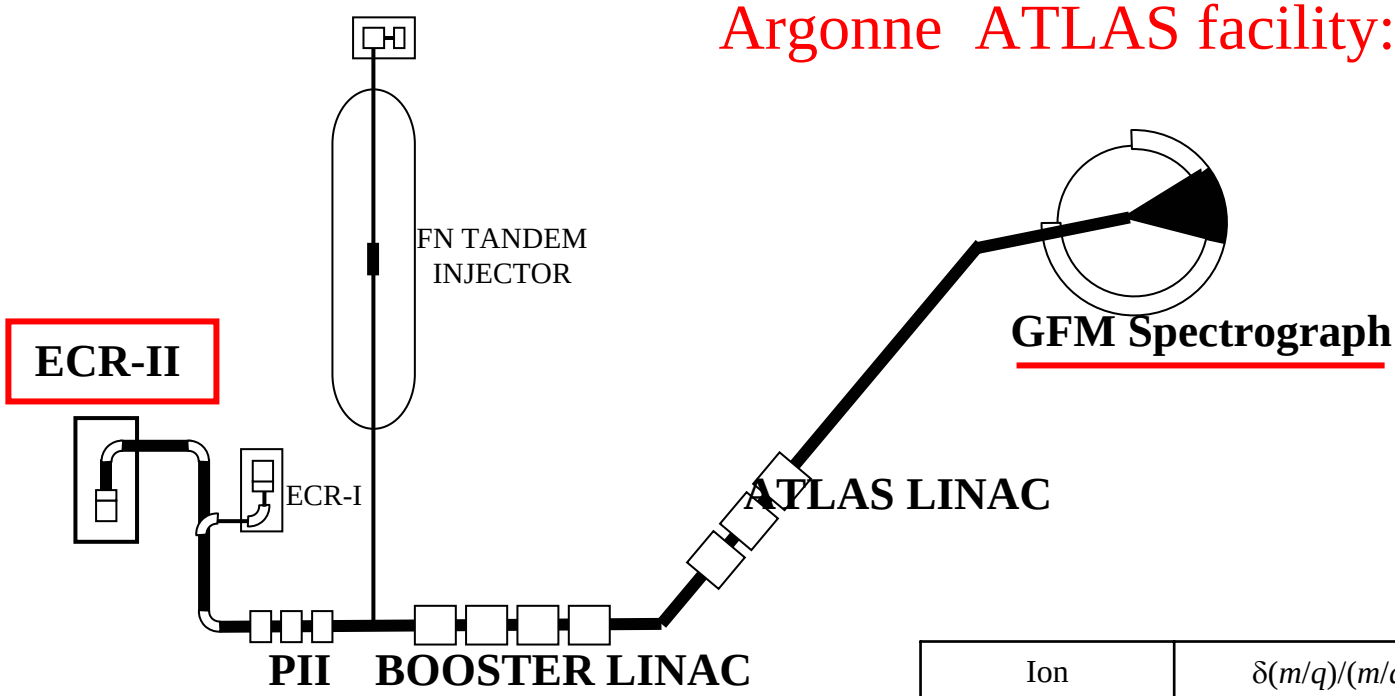
Identification → high energy for discrimination by dE/dx

Argonne ATLAS facility:



Ion	$\delta(m/q)/(m/q)$ %
146Sm22+	0.000
146Nd22+	0.000
152Sm23+	– 0.417
80Kr12+	+ 0.457
133Xe20+	+ 0.205
139La21+	– 0.261
166Er25+	– 0.055

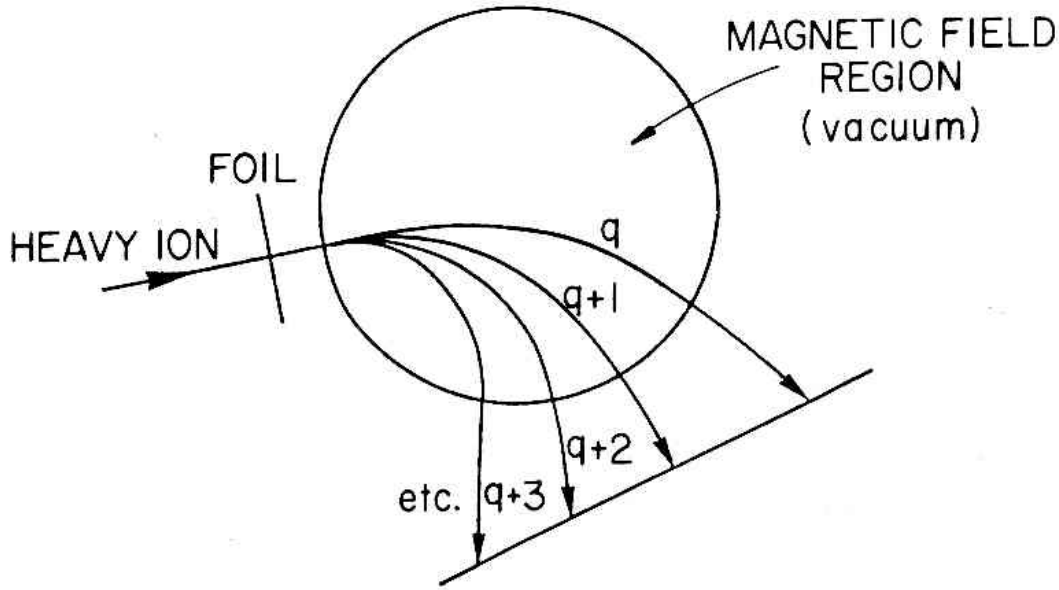
Accelerated Ions: iso-(m/q)

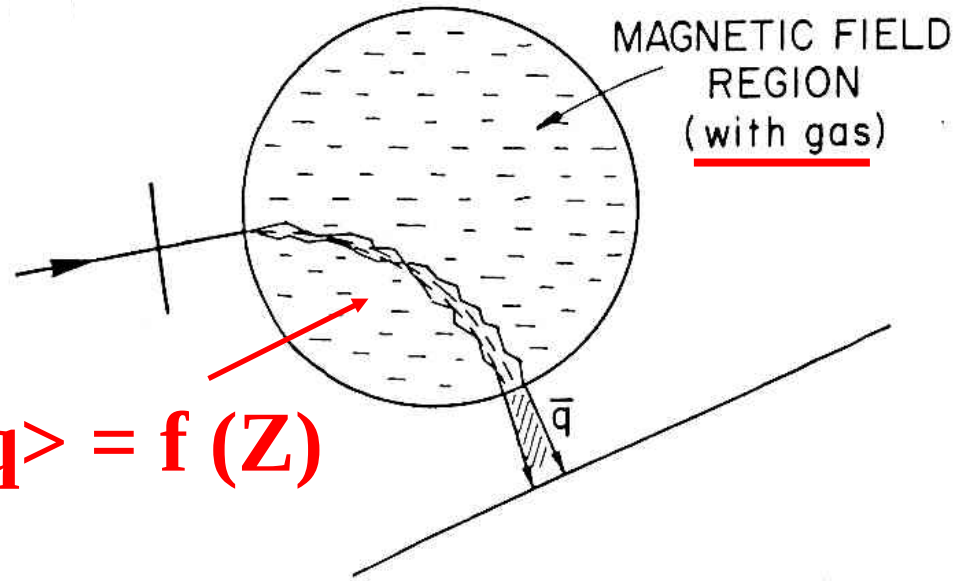
$^{146}Sm^{22+}$, $^{146}Nd^{22+}$

m/q=146/22

Pilot beam : $^{80}Kr^{12+}$, $^{147}Sm^{22+}$

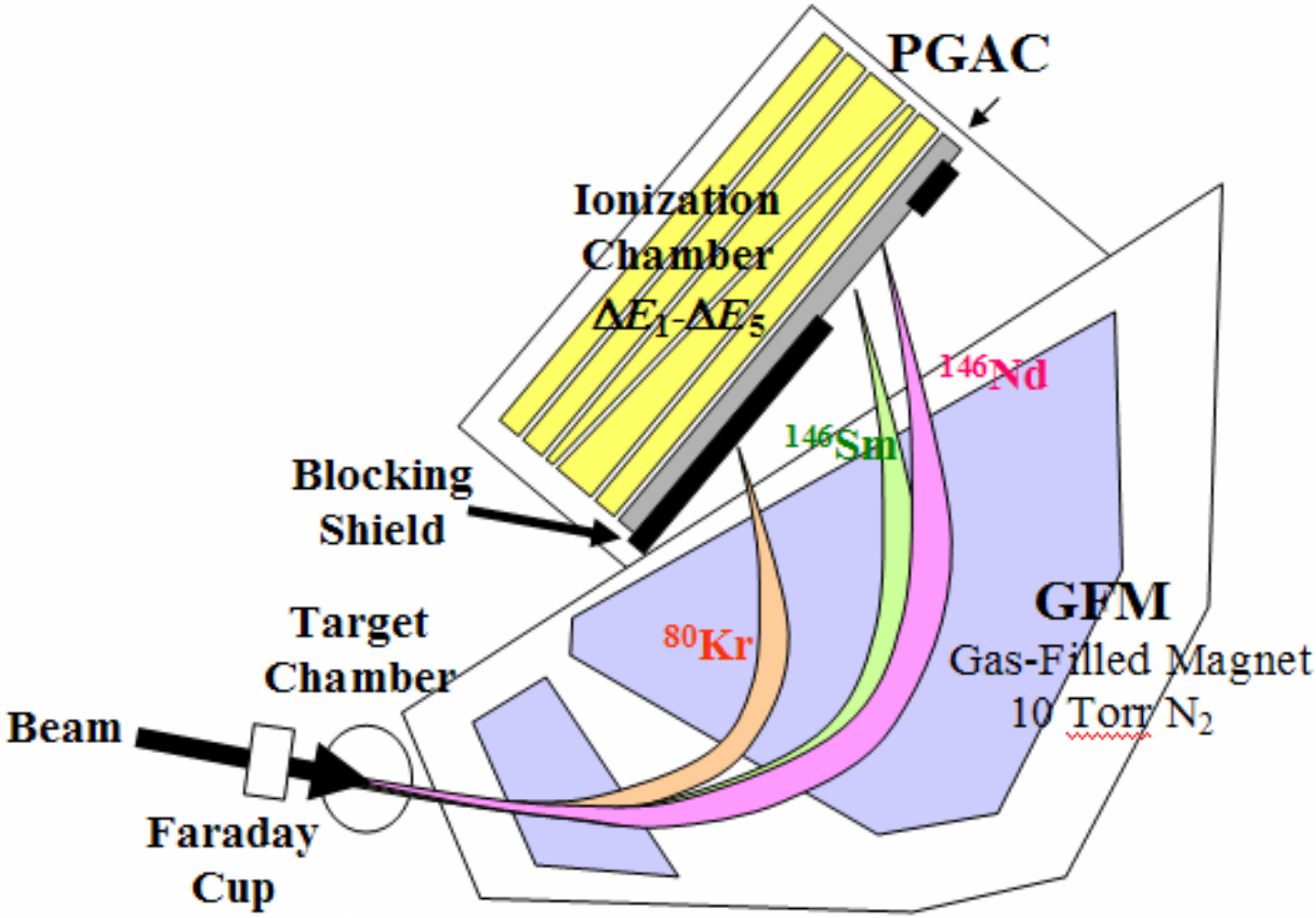
Gas-filled magnetic Spectrograph : principle

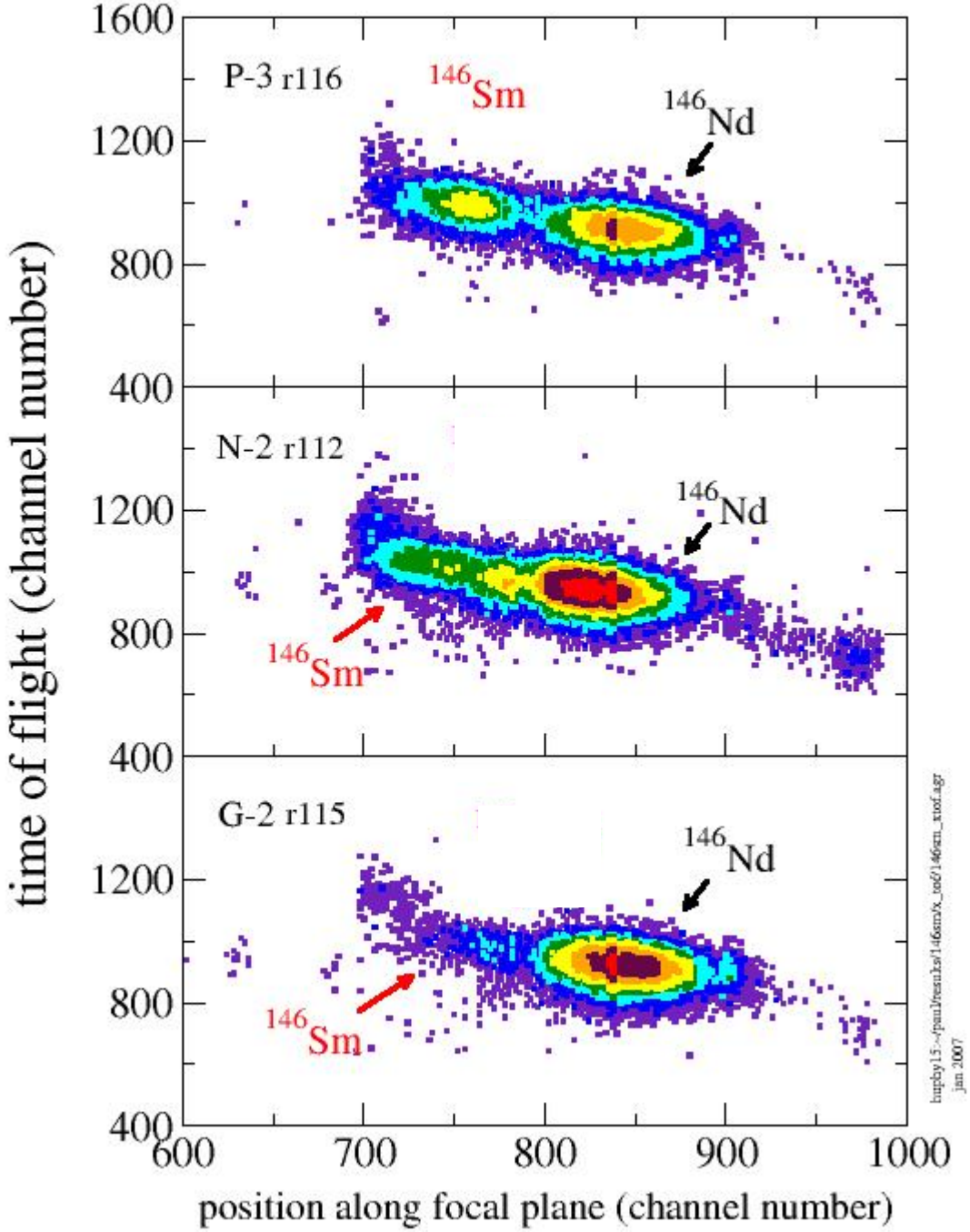


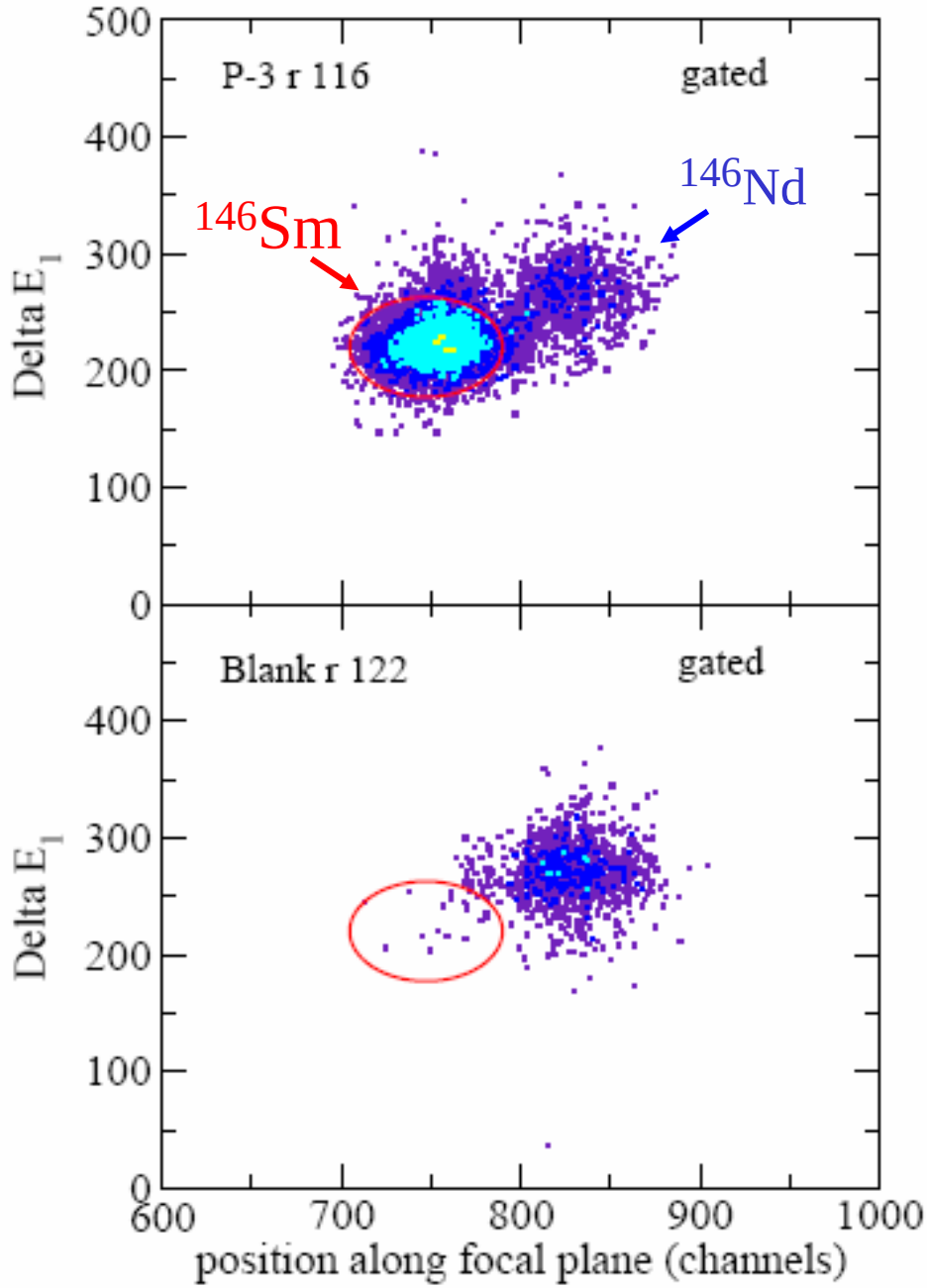


<q> = f (Z)

GFM Spectrograph

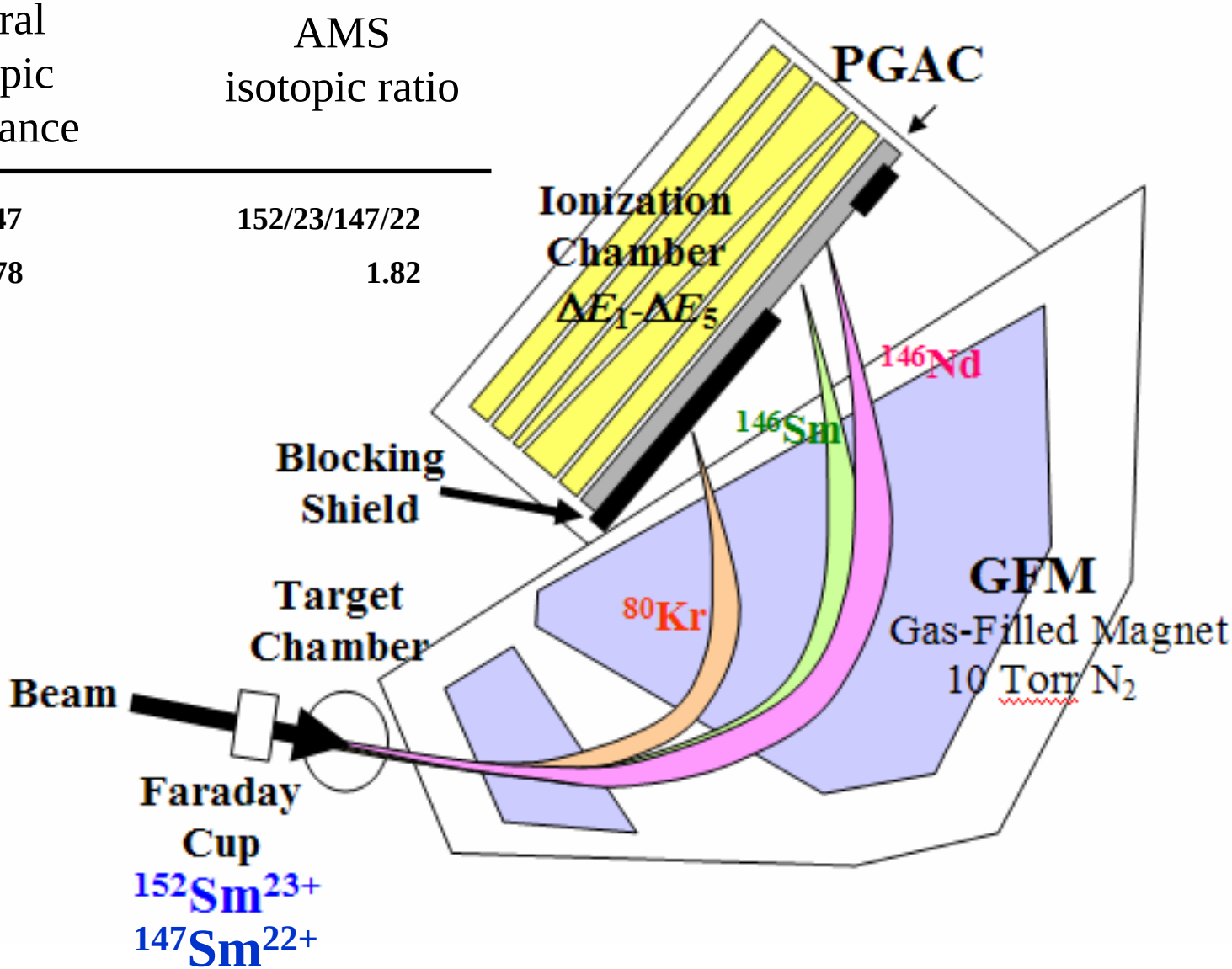



P-3 r116
146Sm
146Nd
N-2 r112
146Nd
146Sm
G-2 r115
146Nd
146Sm
time of flight (channel number)
position along focal plane (channel number)
1600
1200
800
400
600
700
800
900
1000
hupby15:~/paul/resn.ks/146sm/x_tof/146sm_xtof.agr
jan 2007


P-3 r 116
gated
146Sm
146Nd
Blank r 122
gated
Delta E1
Delta E1
500
400
300
200
100
0
400
300
200
100
0
600
700
800
900
1000
position along focal plane (channels)

GFM Spectrograph

natural isotopic abundance	AMS isotopic ratio
152/147	**152/23/147/22**
1.78	**1.82**



Present status summary:

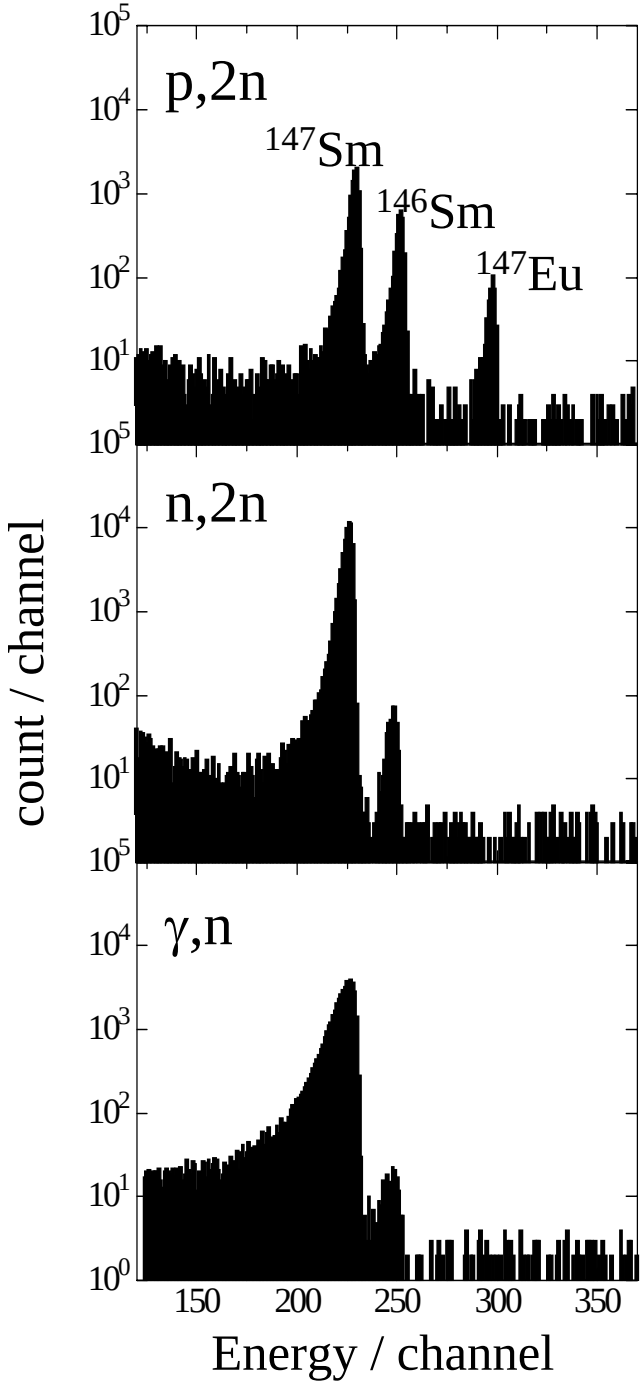
1. good separation and identification of ^{146}Sm by high-energy AMS

2. present sensitivity : $^{146}Sm/Sm \sim 10^{-11}$

3. half-life re-determination in progress : need to measure $^{146}Sm/^{147}Sm$ ratio in absolute terms.

$$A_{146} = \frac{\ln 2}{t_{146}} \times N_{146} \qquad A_{147} = \frac{\ln 2}{t_{147}} \times N_{147}$$

$$t_{146} = \frac{A_{147}}{A_{146}} \times \frac{N_{146}}{N_{147}} \times t_{147}$$


p,2n
147Sm
146Sm
147Eu
n,2n
γ,n
count / channel
Energy / channel
150
200
250
300
350

Present status summary:

1. good separation and identification of ^{146}Sm by high-energy AMS

2. present sensitivity : $^{146}Sm/Sm \sim 10^{-11}$

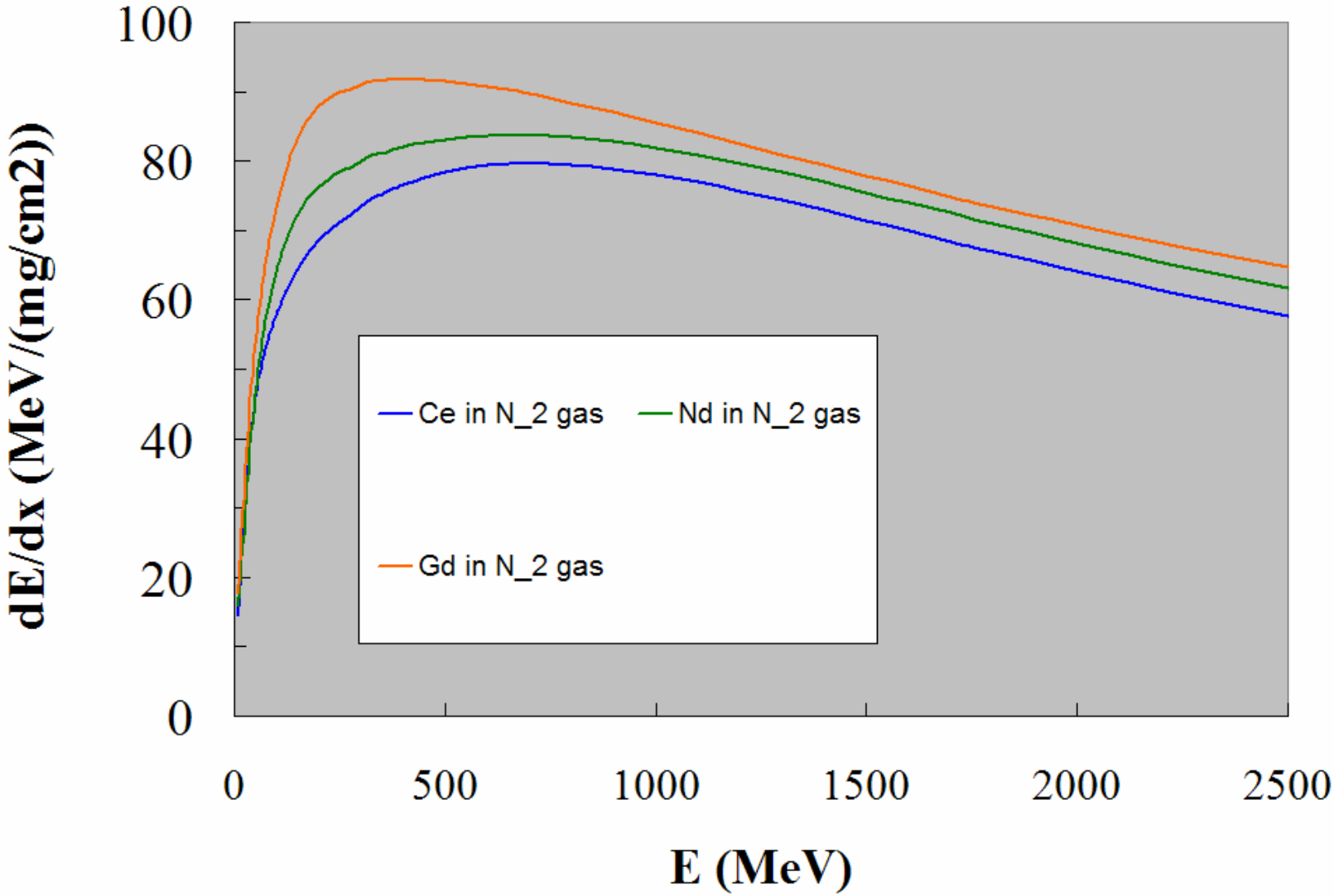
3. half-life re-determination in progress : need to measure $^{146}Sm/^{147}Sm$ ratio in absolute terms.

Half-life (year)	Author
5×10^7	D. C. Dunlavey and G. T. Seaborg, 1953
$7.4 \pm 1.5 \times 10^7$	M. Nurmia et al., 1964
$1.03 \pm 0.05 \times 10^8$	A. M. Friedman et al., 1966
$1.03 \pm 0.05 \times 10^8$	F. Meissner et al., 1987

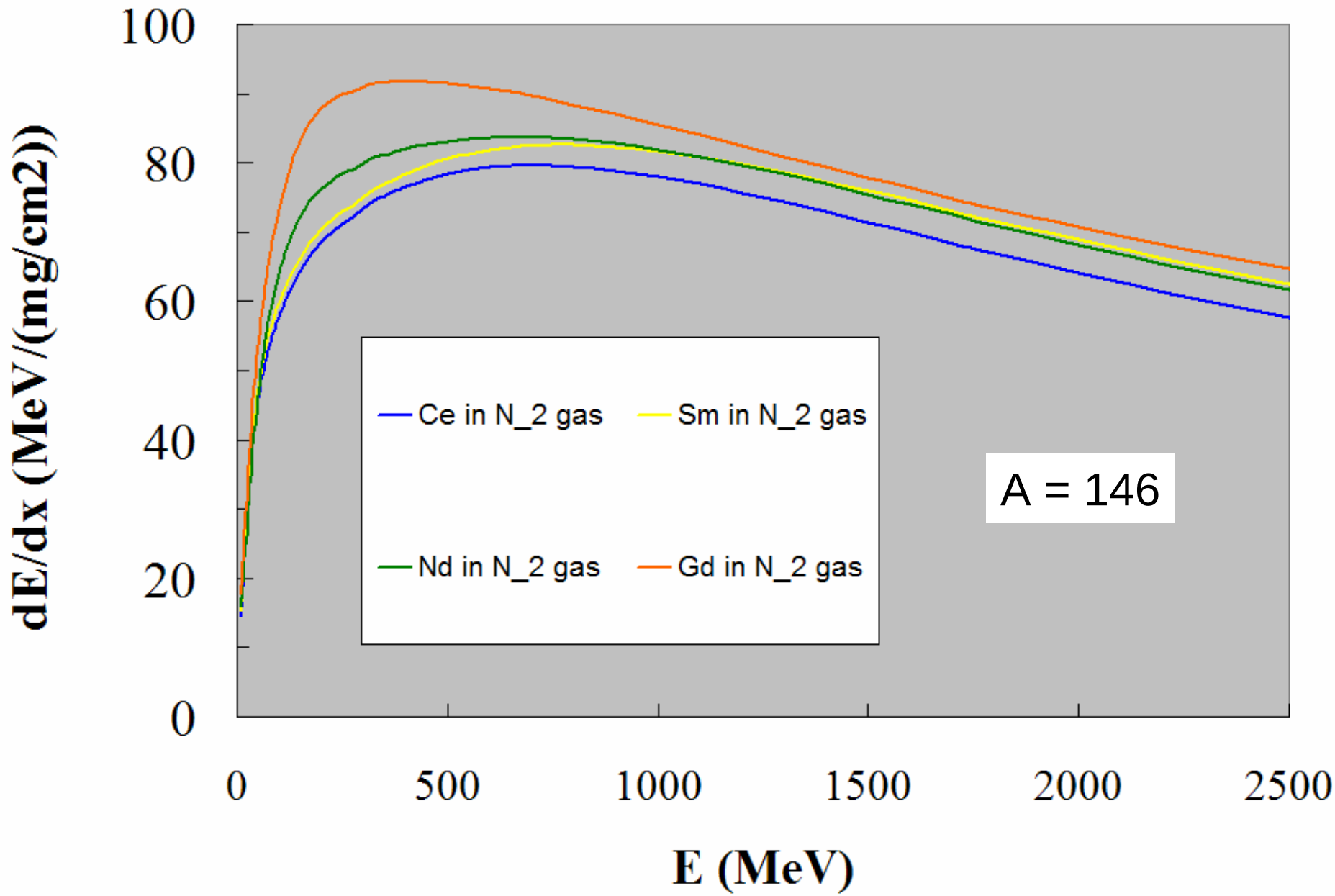
^{146}Sm samples

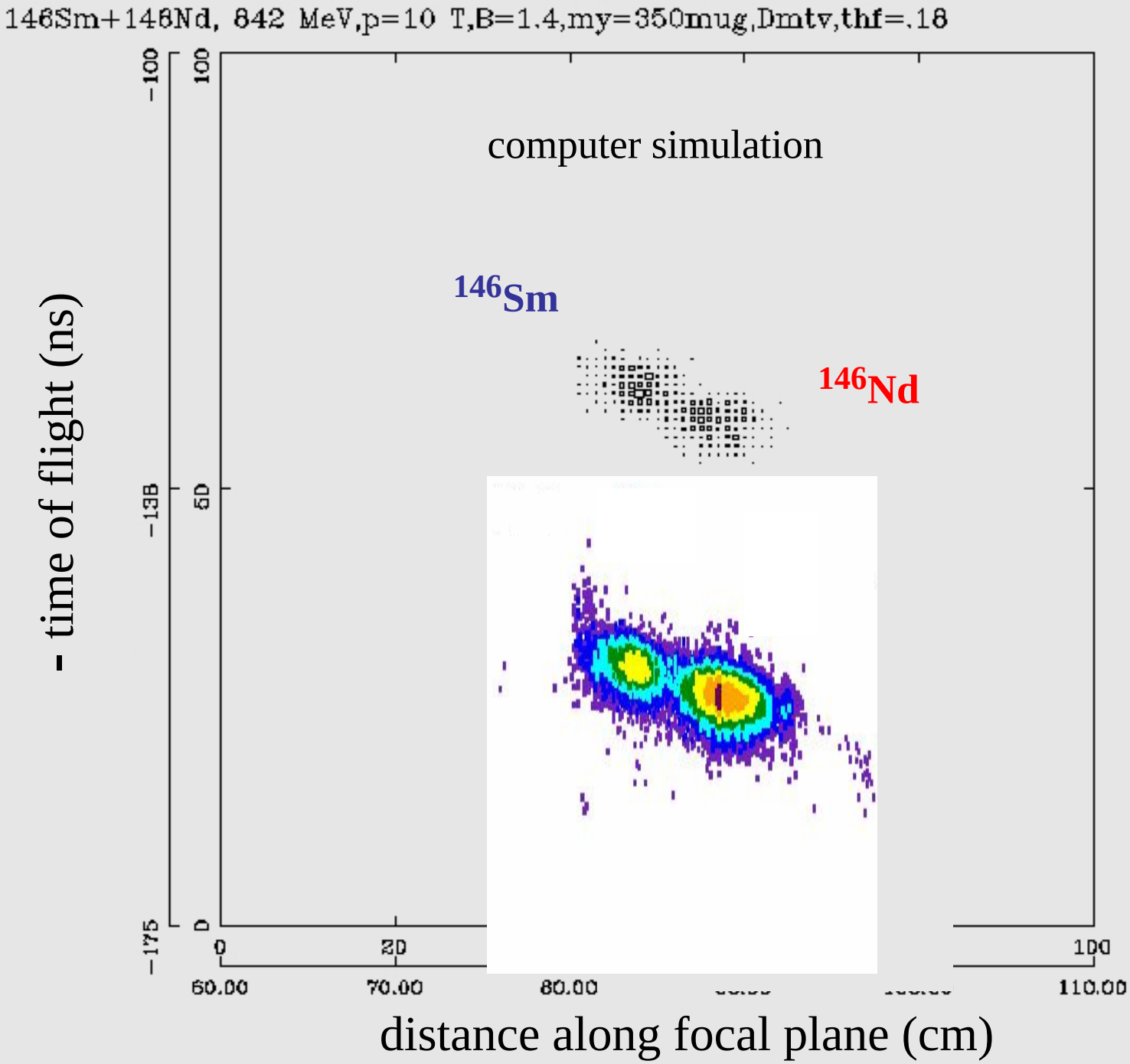
Sample name	146/147 activity ratio (a)	estimated 146/147 atom ratio (a)	^{147}Sm activation
(gamma, n)			
G-1	2.28x10^-7	~2x10^-10	50 MeV electrons Tohoku U.
G-2	2.95x10^-5	~3x10^-8	
(n, 2n)			
N-1	5.18x10^-7	~5x10^-10	fast neutrons Japan Material Testing Reactor
N-2	6.15x10^-5	~6x10^-8	
(p,2n)&EC			
P-1	2.50x10^-7	~3x10^-10	21 MeV protons AVF cyclotron Osaka U.
P-2	2.33x10^-7	~2x10^-10	
P-3	5.45x10^-4	~5x10^-7	
P-4	5.12x10^-4	~5x10^-7	
blank: natural Sm used for dilution			
B	0.00	0.00	

(a): after dilution of activated material with ^{147}Sm and Sm purification. Samples were precipitated and ignited to Sm_2O_3. Final masses of samples ~ 50 mg.

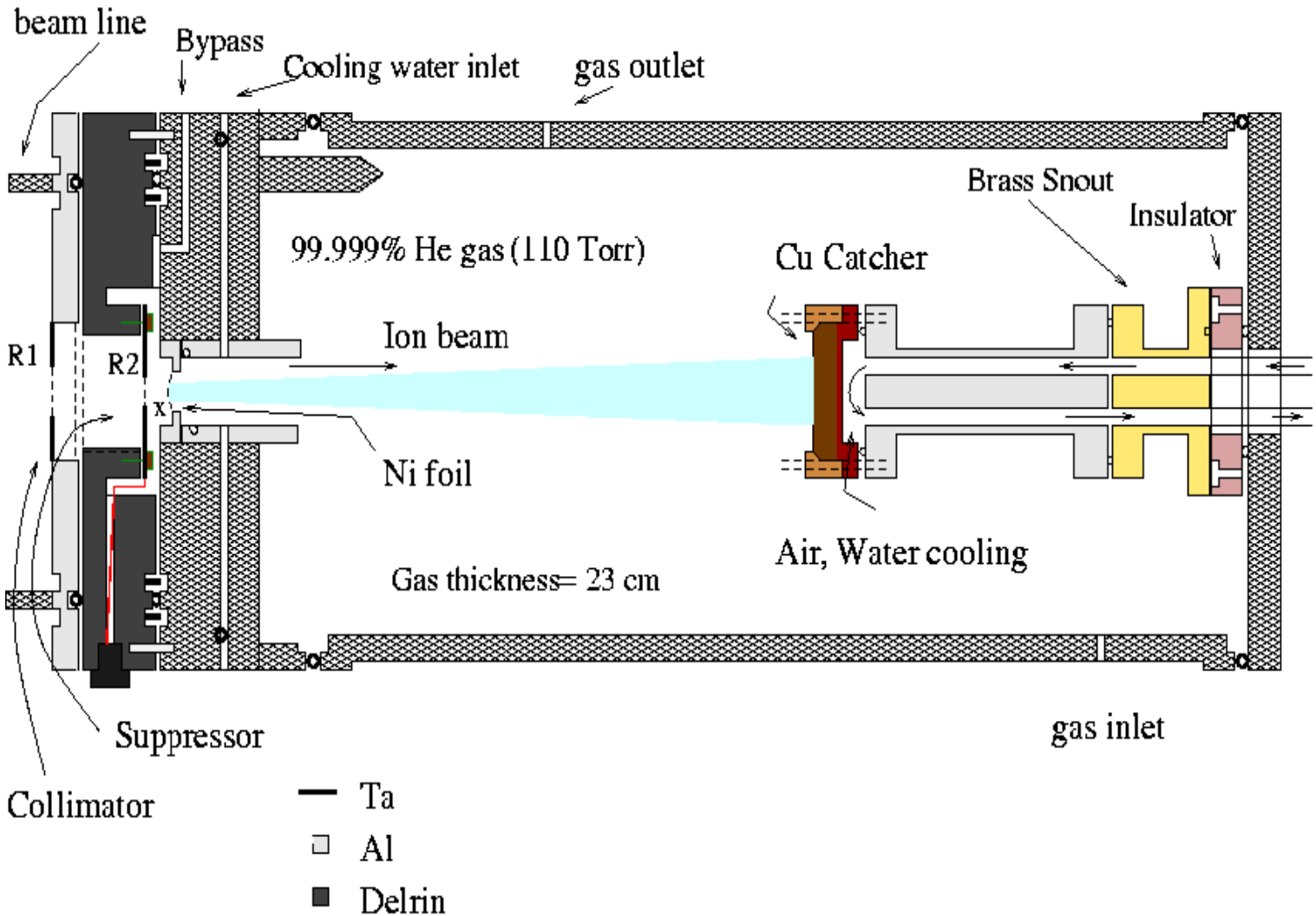

100
80
60
40
20
0
dE/dx (MeV/(mg/cm2))
0
500
1000
1500
2000
2500
E (MeV)
Ce in N_2 gas
Nd in N_2 gas
Gd in N_2 gas

SRIM 2006


100
80
60
40
20
0
dE/dx (MeV/(mg/cm2))
0
500
1000
1500
2000
2500
E (MeV)
Ce in N_2 gas
Sm in N_2 gas
Nd in N_2 gas
Gd in N_2 gas
A = 146


146Sm+146Nd, 842 MeV,p=10 T,B=1.4,my=350mug,Dmtv,thf=.18
computer simulation
146Sm
146Nd
- time of flight (ns)
distance along focal plane (cm)
-100
100
-138
50
-175
0
0
20
100
60.00
70.00
80.00
110.00

target chamber & Cu catcher assembly



S.B. Jacobsen, G.J. Wasserburg, Earth Planet. Sci. Lett. 67, 137 (1984):
^{146}Sm in-situ decay in meteorites : ^{146}Sm present in Early-Solar system

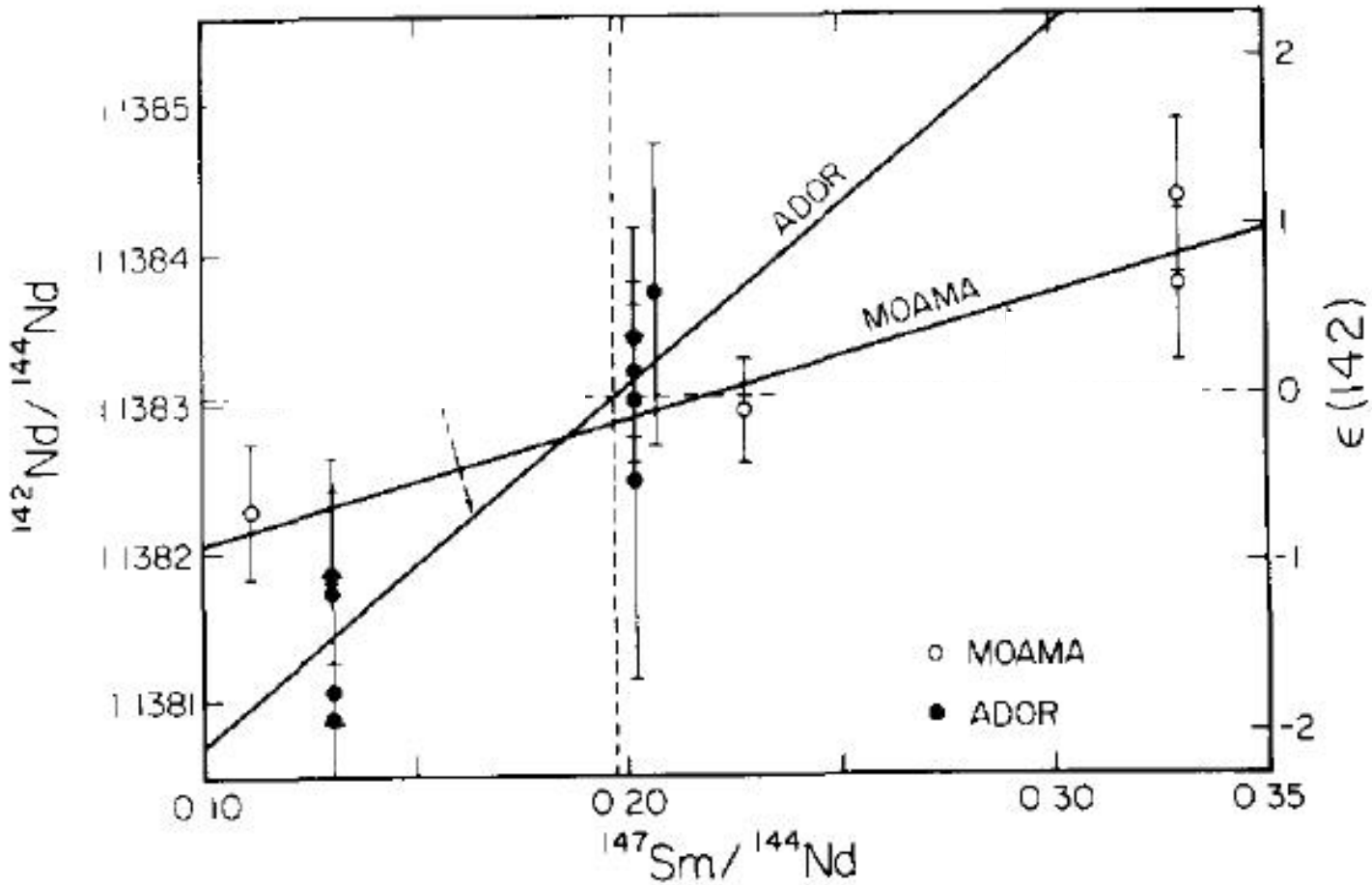


Fig 6 ^{142}Nd/^{144}Nd evolution diagram for the achondrites Moama and Angra dos Reis.

E. Somorjai *et al*., Astron. and Astroph. 333, 1112 (1998)

$^{144}Sm(\alpha,\gamma)^{148}Gd$

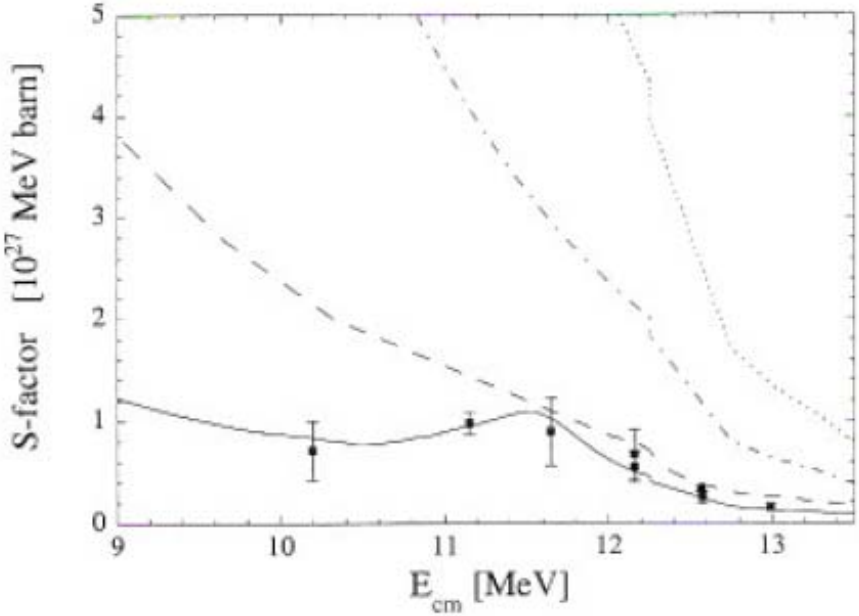


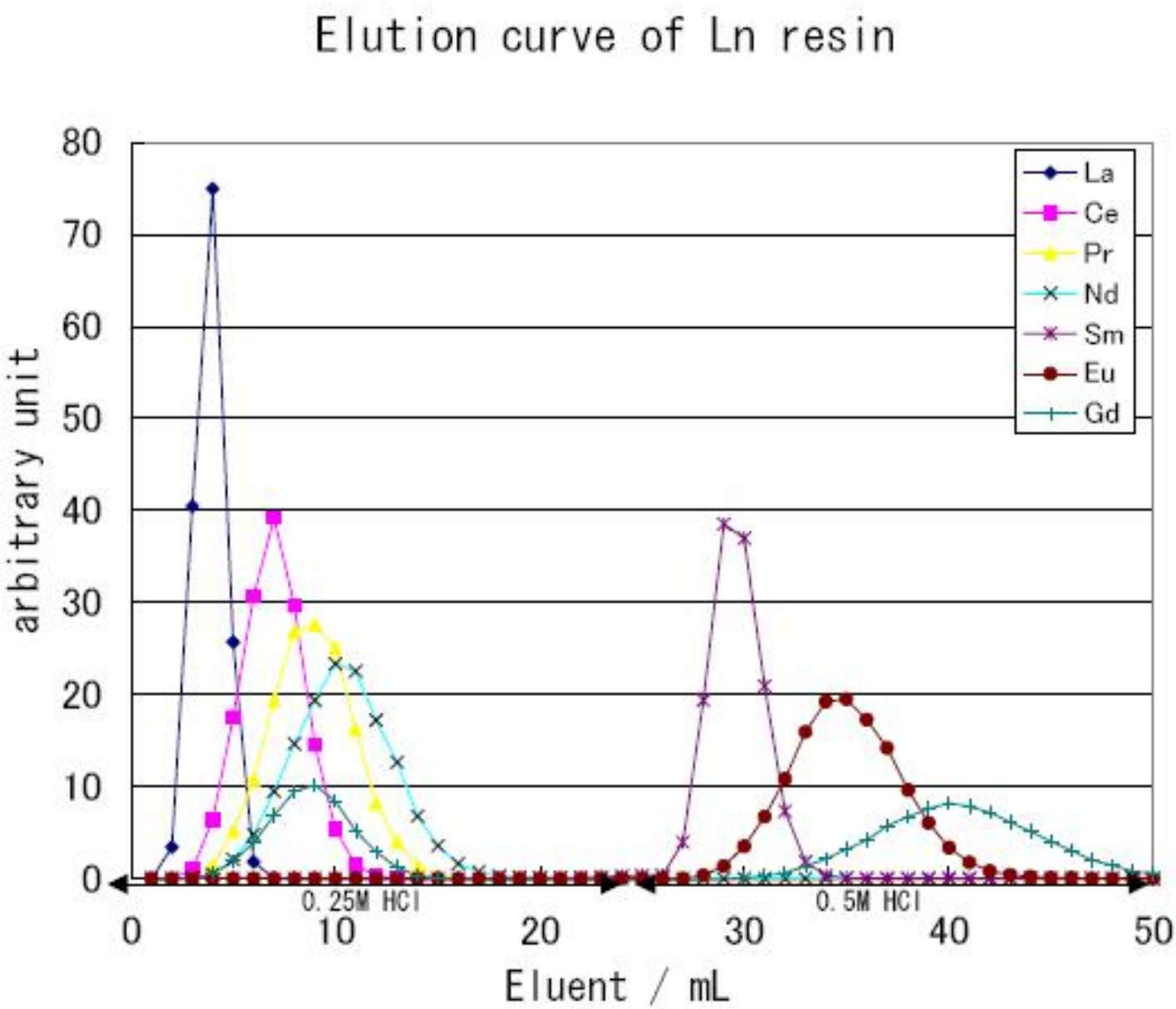
Table 1. Experimental cross sections and S-factors for $^{144}Sm(\alpha,\gamma)^{148}Gd$

E_α [a)] MeV	$E_{cm}^{eff.}$ [b)] MeV	σ μbarn	$\Delta\sigma$ μbarn	$S(\Delta S)$ 10^{26} MeV-barn
10.500	10.193	0.083	0.034	7.13 (2.92)
11.500	11.151	2.9	0.3	9.76 (1.01)
12.000	11.647	12.1	4.5	8.94 (3.32)
12.509	12.156	39.2	14.1	6.74 (2.42)
12.505	12.159	32.4	9.0	5.52 (1.53)
12.939	12.571	45.7	11.0	2.55 (0.61)
12.942	12.568	59.0	5.9	3.33 (0.33)
13.355	12.992	83.2	16.6	1.58 (0.31)

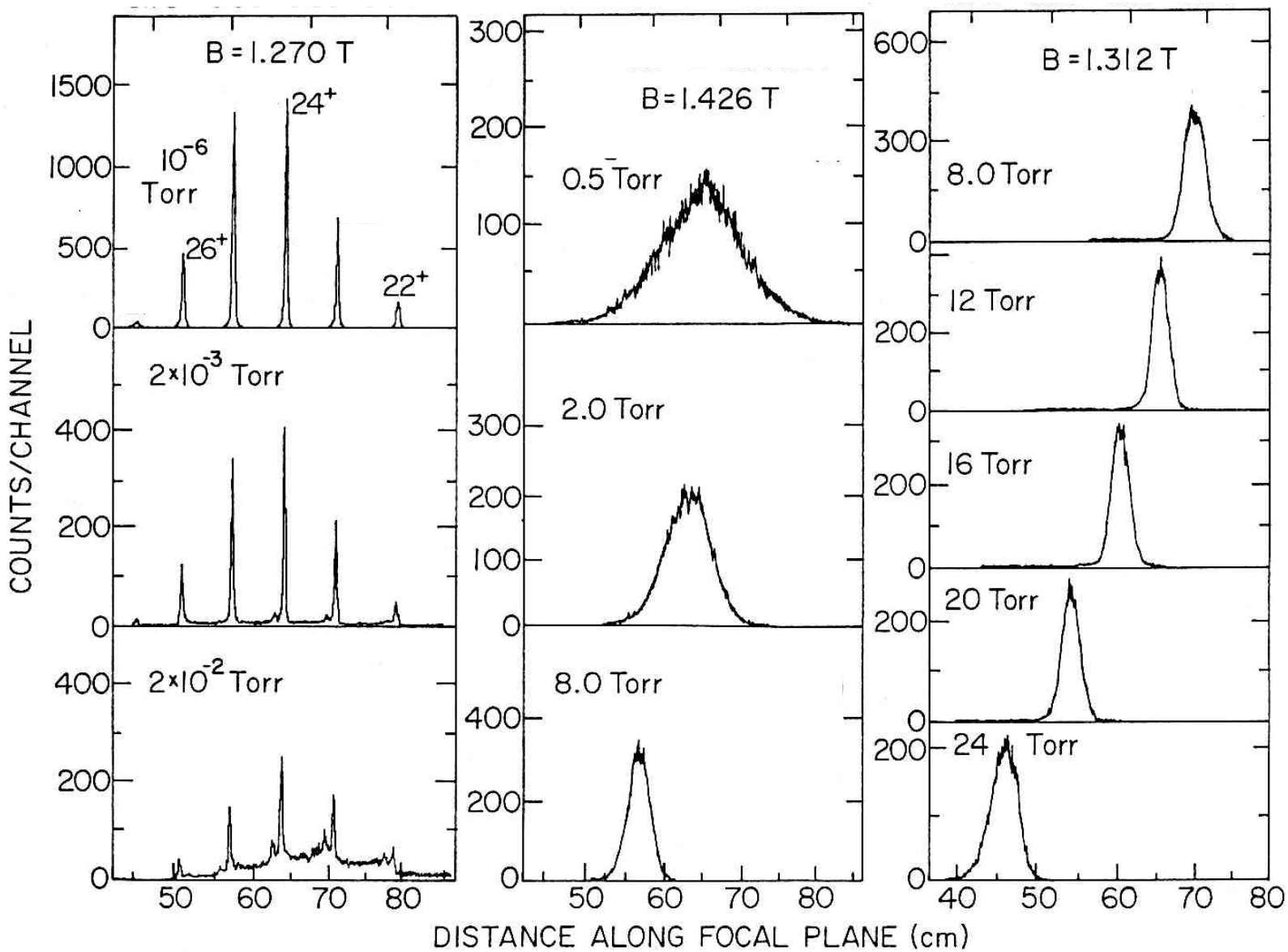
a) errors for $E_\alpha \leq 12$ MeV and for others are $\pm$ 1.5 keV and $\pm$ 7 keV (cyclotron), respectively

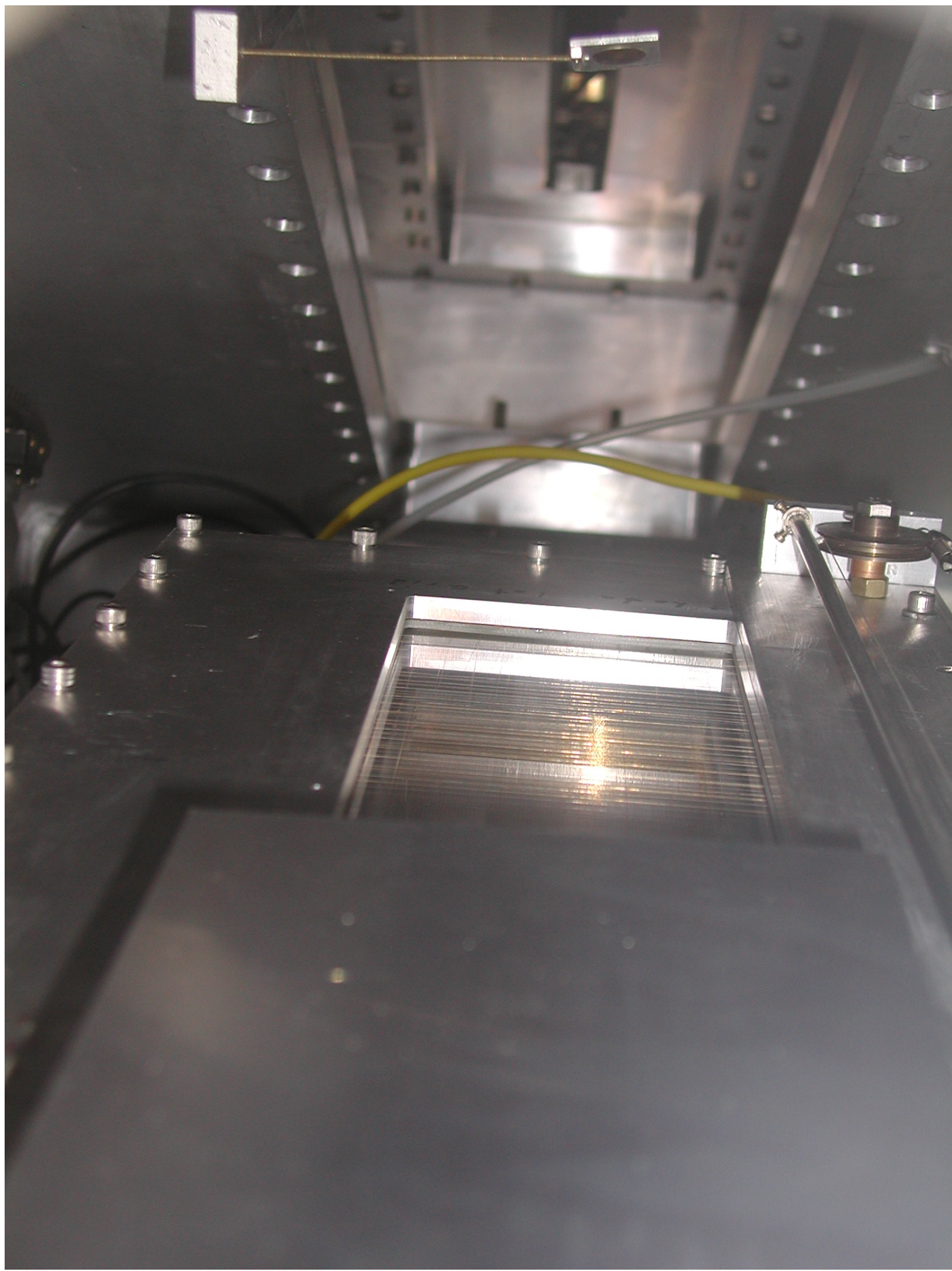
b) calculated from the target thicknesses

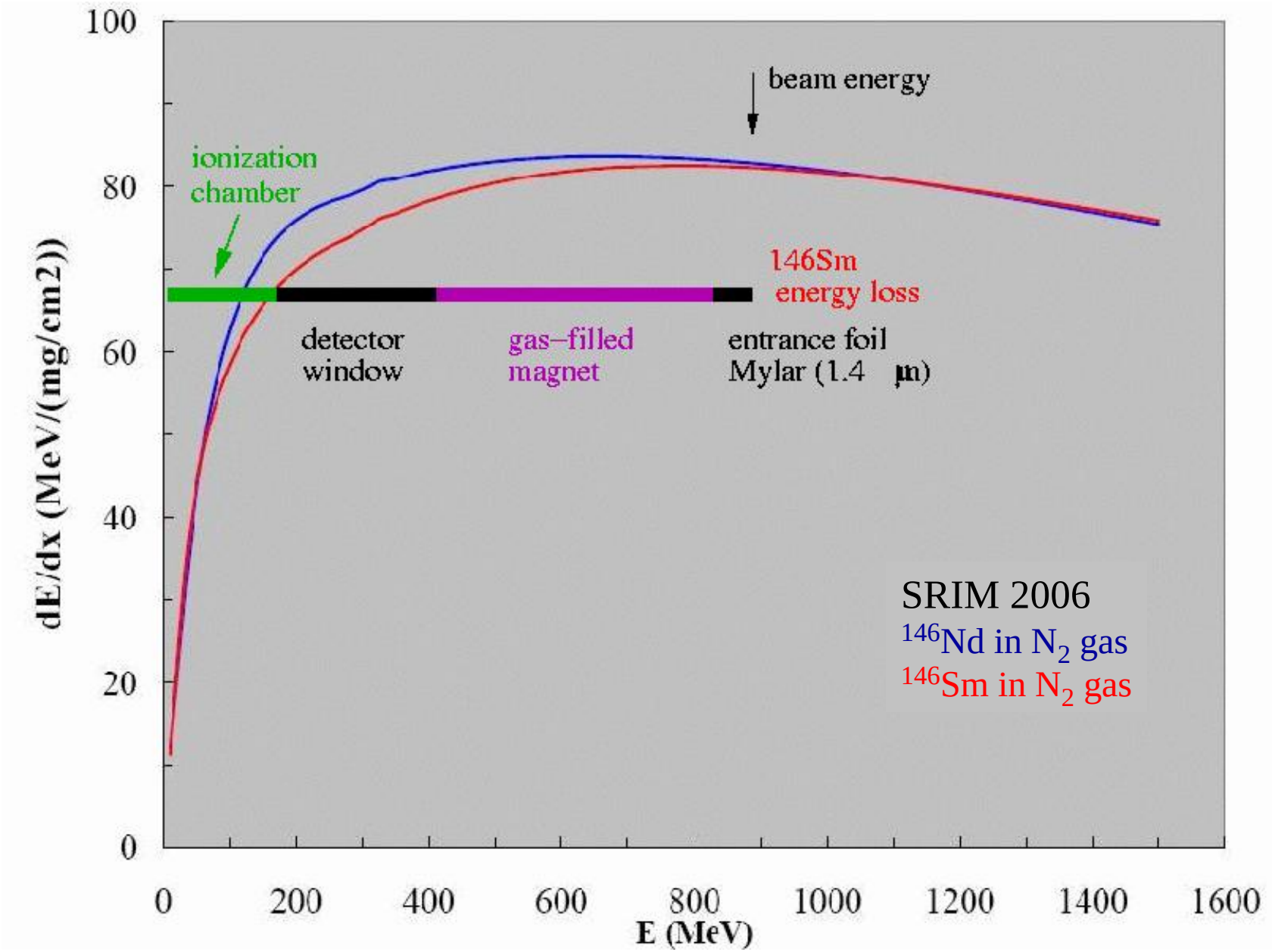
e.g.: $^{142}Nd\ (\alpha,\gamma)\ ^{146}Sm$, measured by counting ^{146}Sm nuclei ?


Elution curve of Ln resin
arbitrary unit
80
70
60
50
40
30
20
10
0
La
Ce
Pr
Nd
Sm
Eu
Gd
0.25M HCl
0.5M HCl
0
10
20
30
40
50
Eluent / mL

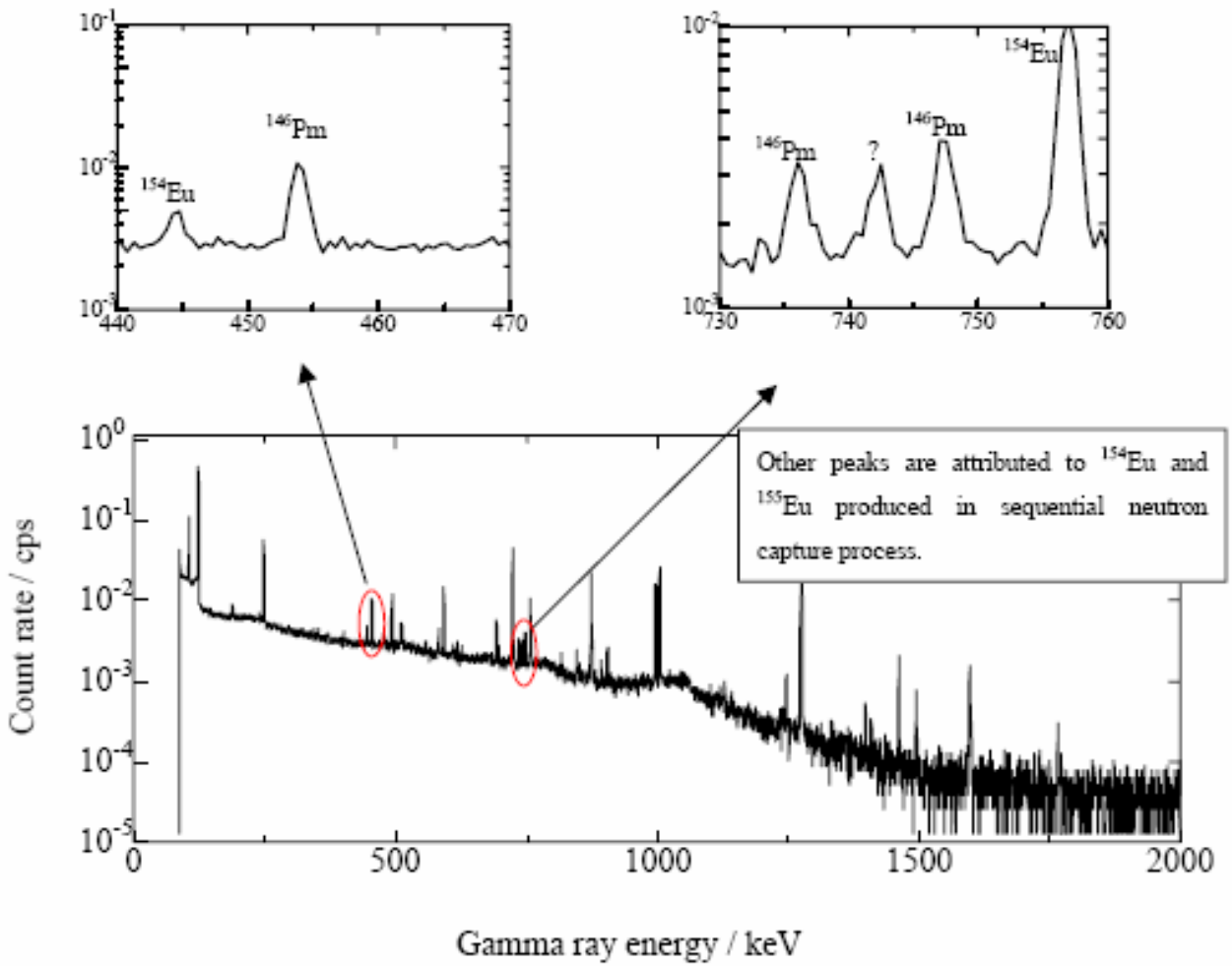
^{58}Ni (350 MeV)


B = 1.270 T
10^{-6} Torr
26^+
24^+
22^+
2×10^{-3} Torr
2×10^{-2} Torr
B = 1.426 T
0.5 Torr
2.0 Torr
8.0 Torr
B = 1.312 T
8.0 Torr
12 Torr
16 Torr
20 Torr
24 Torr
COUNTS/CHANNEL
DISTANCE ALONG FOCAL PLANE (cm)

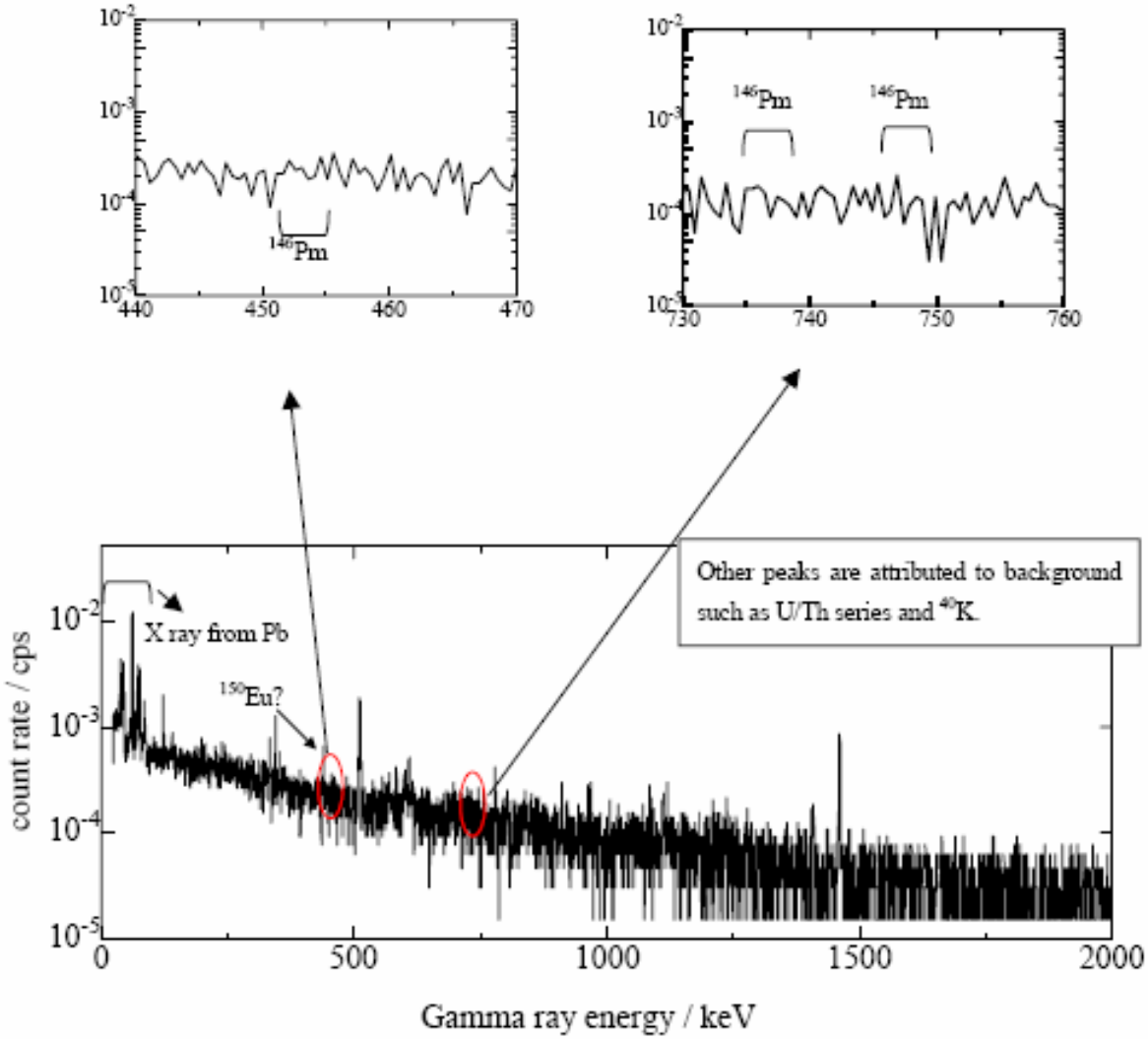


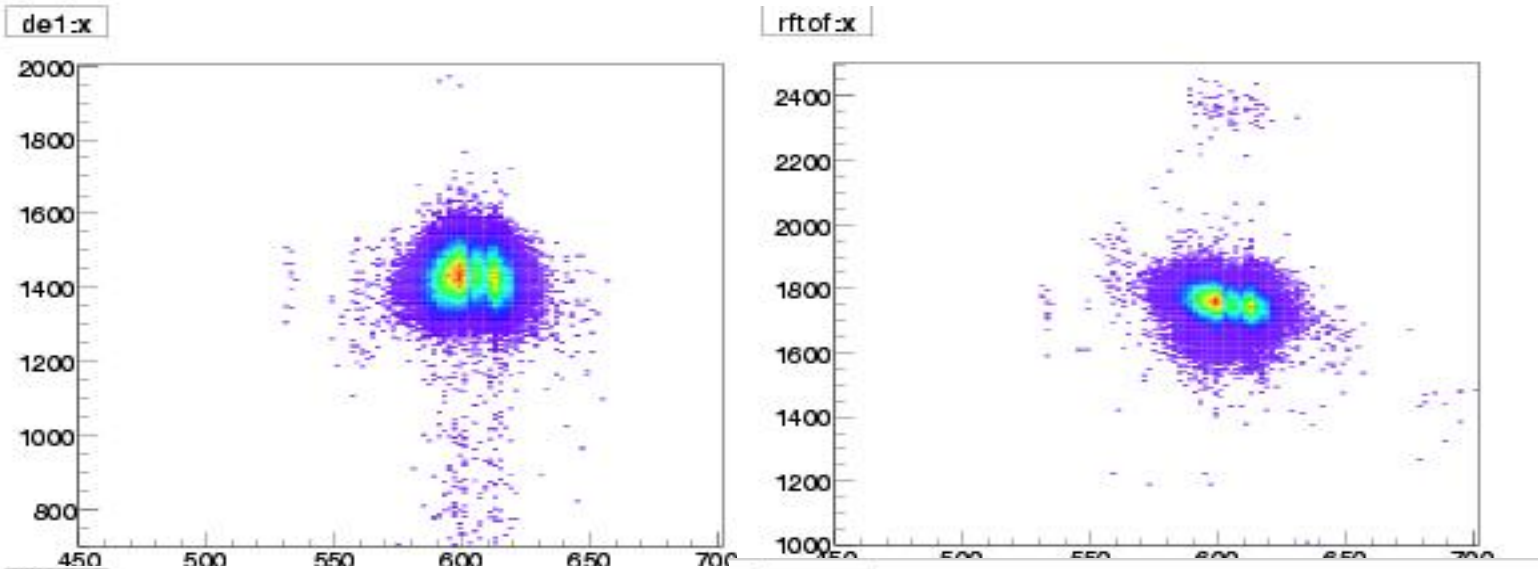

100
80
60
40
20
0
dE/dx (MeV/(mg/cm2))
0
200
400
600
800
1000
1200
1400
1600
E (MeV)
beam energy
ionization
chamber
146Sm
energy loss
detector
window
gas–filled
magnet
entrance foil
Mylar (1.4 μm)
SRIM 2006
146Nd in N2 gas
146Sm in N2 gas

Neutron Activated ^{146}Sm sample

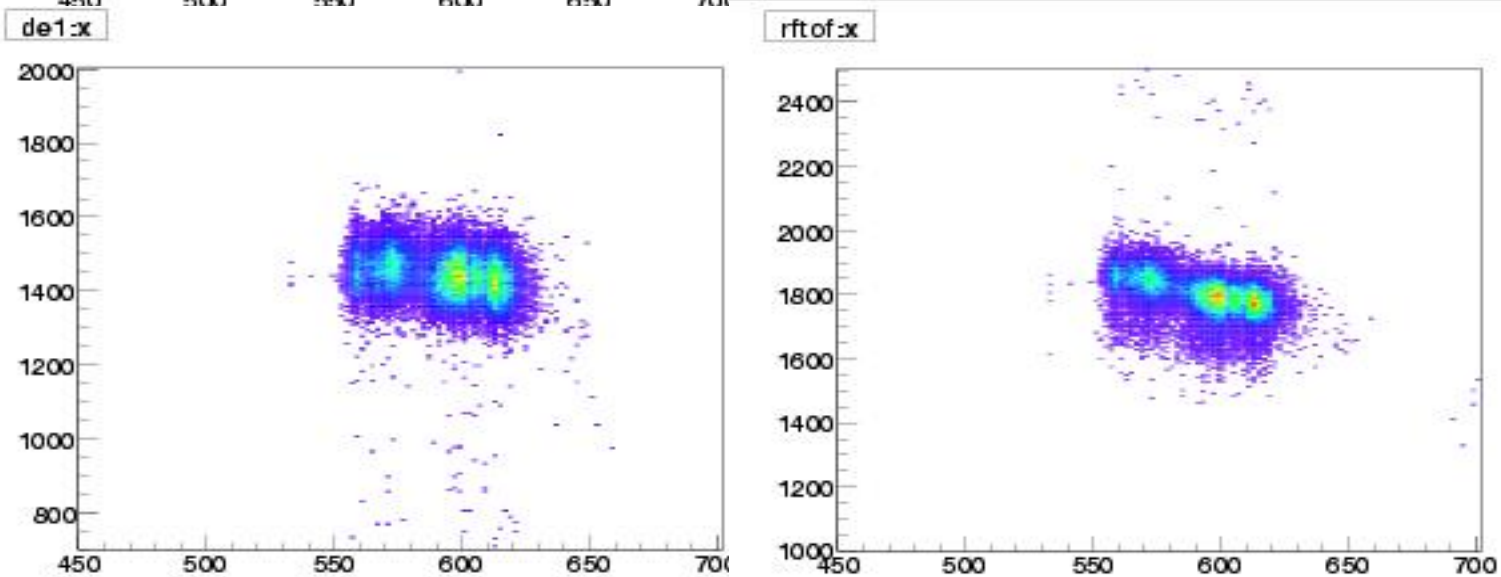


Proton Activated ^{146}Sm sample





blank sample



P-4 sample